IAEA-SM-254/63
CONF-810655--1

DEVELOPMENT AND APPLICATION OF TERRESTRIAL FOOD CHAIN MODELS TO ASSESS HEALTH RISKS TO MAN AND RELEASES OF POLLUTANTS TO THE ENVIRONMENT*

Stephen V. Kaye, F. Owen Hoffman,
Laura M. McDowell-Boyer, and C. Fred Jaes

Health and Safety Research Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

MASTER

ABSTRACT

This paper reviews the development and application of mathematical models used to predict the terrestrial food chain transport of pollutants of potential importance to human health. A distinction is made between models developed specifically for assessment applications and models which may function as research tools. Differentiation is also made between models whose structure is based on steady-state relationships among food chain compartments and dynamic models developed to simulate food chain and pollutant kinetics. The strengths and weaknesses of these models are related to the needs of the model user, the availability of relevant data for parameter quantification, and the feasibility for model validation. For assessment purposes, an optimum level of structural complexity will be achieved when all parameters are readily measurable and predictive error due to unforeseen correlations among parameters is small. The optimum level of simplification, however, will be determined by model validation results and the ease of model implementation. Most examples are derived from models used to assess the terrestrial food chain transport of radionuclides because assessment methodologies for other types of pollutants are only at an early stage of development. It is concluded that current limitations in parameter quantification and model validation will probably restrict most assessment applications of terrestrial food chain models to a type of screening calculation. However, once pollutant releases actually occur, environmental monitoring will be necessary to ensure that potential model misprediction does not result in unacceptable consequences.

By acceptance of this article, the publisher or recipient acknowledges the U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering the article.

*Research sponsored by the U. S. Department of Energy under contract W-7405-eng-26 with the Union Carbide Corporation.

DISCLAIMER

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MGW

1. INTRODUCTION

Terrestrial food chains, because of their dietary inputs to human populations, are of particular importance in the assessment of health impact from pollutant releases to the environment. Transport through food chains may result in pollutant concentrations which exceed background or natural levels and thus increase the risk to human health. Although transport and bioaccumulation of pollutants occurs in aquatic as well as terrestrial systems, our attention is focused on the terrestrial environment because 1) the atmosphere is a major recipient and disperser of pollutant releases, 2) the application of toxic chemicals directly to terrestrial food products for pest control is wide spread, and 3) a major portion of the human diet is derived from agriculture.

Terrestrial food chain transport starts primarily with reactive gases or particulates that are readily deposited on and retained by vegetation. The deposition of potentially soluble pollutants onto soil may also result in food chain contamination through incorporation of these pollutants into edible plant tissue through root uptake. Even for relatively insoluble pollutants, the soil may function as a secondary source of pollutants in terrestrial food chains long after direct releases to the atmosphere or water have ceased (Russell 1966, Garner 1971, Lichtenstein 1972, Edwards 1973). When vegetation contamination has occurred, pollutants may then be available for direct human consumption or transported to food products derived from grazing animals.

For the assessment of environmental or human health impacts, prediction of food chain transport is essential. Food chain transport will not only affect the estimate of health risk to man, but it will also influence the assessment of compliance with regulatory standards and the determination of

the relative importance of various pollutants. The determination of the relative importance of pollutants is especially critical to the evaluation of pollution abatement strategies, including site selection and decisions concerning alternative technologies.

One very useful technique for assessing the consequences of food chain transport of pollutants is through the development and application of mathematical models. Figure 1 represents a conceptual terrestrial food chain model. A mathematical model quantifies the components of the food chain and their interactions. In such a model, food chain components are referred to as "compartments" or "state variables" and the interactions as "transfer coefficients." The transfer coefficients and any scaling factors which relate them to the state variables are referred to as model parameters. In a mathematical model the amount of a pollutant in one compartment may be predicted by defining the relationships between this compartment and all other model compartments and by defining the amount of pollutant input to the system. This predictive capability makes mathematical models ideal tools for decision making in the assessment of pollutant releases to the environment.

At Oak Ridge National Laboratory we have obtained a considerable depth of experience with both the development and application of terrestrial food chain models. The purpose of this paper, therefore, will be to review, within the framework of our experience, the types of models used in the assessment of nuclear and chemical pollutants. We will also discuss the strengths and weaknesses of these models and will recommend directions for future research to improve application of the models and to increase confidence in model predictions A special emphasis will be placed on recommendations for the application and refinement of current models in situations, such as in developing countries, where available data are severely limited.

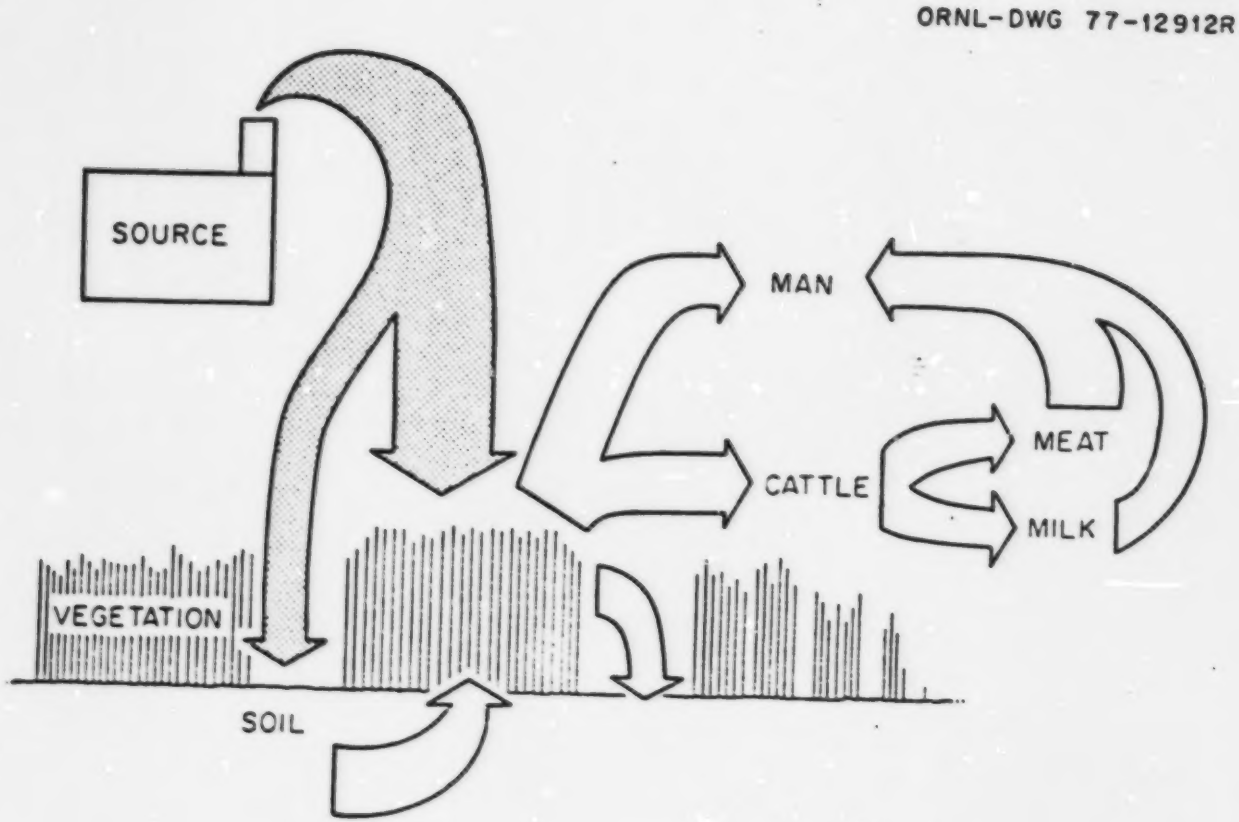


Figure 1. A conceptual terrestrial food chain model for the assessment of health risks to man and releases of pollutants to the environment (from Garten 1978).

2. TYPES OF MODELS

2.1 Model Categories

Although various classifications of models are possible, we recognize two basic categories of models developed to study the fate of pollutants in terrestrial systems: assessment models and research models.

2.1.1 Assessment Models

The primary purpose for assessment models is to predict environmental concentrations of pollutants and associated health risks and thus serve as tools for decision making. In the absence of detailed data, assessment models may focus on only a few basic food types such as cow milk, beef, and leafy vegetables which comprise a major portion of the human diet and are considered major pathways of transport. In these models, underlying processes and mechanisms may be aggregated into general transfer coefficients that relate the concentration of a pollutant in one compartment of a food chain to another.

The predictions made by assessment models must be defensible. The model should accurately describe the system and be based on the most current knowledge of the system that it represents. Parameter values and model predictions should be capable of being validated through field testing. In the absence of results obtained through field validation, assessment models are often given an intentional degree of conservative bias to reduce the probability of underestimation. As will be discussed later, such a conservative bias is most useful for calculations designed to screen for pathways and pollutants that may be of potential importance to human health, regulatory standards, or effluent design objectives.

2.1.2 Research Models

Research models often differ from assessment models in their objectives. Emphasis is often placed on the ability of the research model to simulate the processes affecting the behavior of the pollutant in the food chain, rather than on the prediction of concentrations in foods of importance to humans. Thus, research models are usually developed at the process level, making them far more complex than is typical of assessment models.

The ability to field test process-level parameters and model predictions is not as important a constraint for the development of research models. For the purposes of research, no efforts are made to include an intentionally conservative bias in model predictions. The development of research models may also not be constrained by the limitations of potential users. Therefore, adaptation of research models to assessments will usually be restricted to specific situations where assessment models do not exist or are not applicable, or when data are unavailable for quantification of key assessment model parameters.

2.2 Model Approaches

A variety of approaches can be taken in the development of terrestrial food chain models. Basically, these approaches can be classified into two general types of models: quasi-equilibrium models and dynamic models. The quasi-equilibrium model is derived from the approach defined by the International Commission for Radiological Protection (ICRP 1979) as the "Concentration Factor (CF) Method" for the assessment of radionuclides, or the "environmental commitment concept" for the assessment of environmental mercury (Barry 1979). These approaches are based on steady-state relationships between food chain compartments. Dynamic models on the other hand

are based on system kinetics. These models can be designed to explicitly address the time-dependent behavior of pollutants in terrestrial food chains. The approach represented by dynamic models is referred to by the ICRP as the "Systems Analysis (SA) Method."

2.2.1 Quasi-Equilibrium Models

Quasi-equilibrium models assume that the concentration of pollutants in some compartments of terrestrial food chains are at steady state and concentrations in other compartments are changing with time. Maximum values occur either at the time of harvest or upon some pre-determined length of exposure of terrestrial system to a continuously uniform or average rate of pollutant deposition. In an equilibrium situation, the sum of the rates of pollutants entering a compartment are equal to the sum of the rates of pollutants leaving the compartment, i.e., the concentration of the pollutant in the compartment of reference does not change with time (Fig. 2). Thus, the concentration in a given food product at equilibrium is determined through simple multiplication, whereby:

$$C_B^* = C_A^* \times P_{AB} \qquad (1)$$

where

C_B^* = the equilibrium concentration of a pollutant in a receptor food chain compartment B,

C_A^* = the steady-state concentration in a donor food chain compartment A, and

P_{AB} = the equilibrium concentration factor for the flow of pollutants between the donor A and receptor B food chain compartments.

The quasi-equilibrium model assumes that some compartments of the model may not be at equilibrium; therefore, the concentrations in these compartments

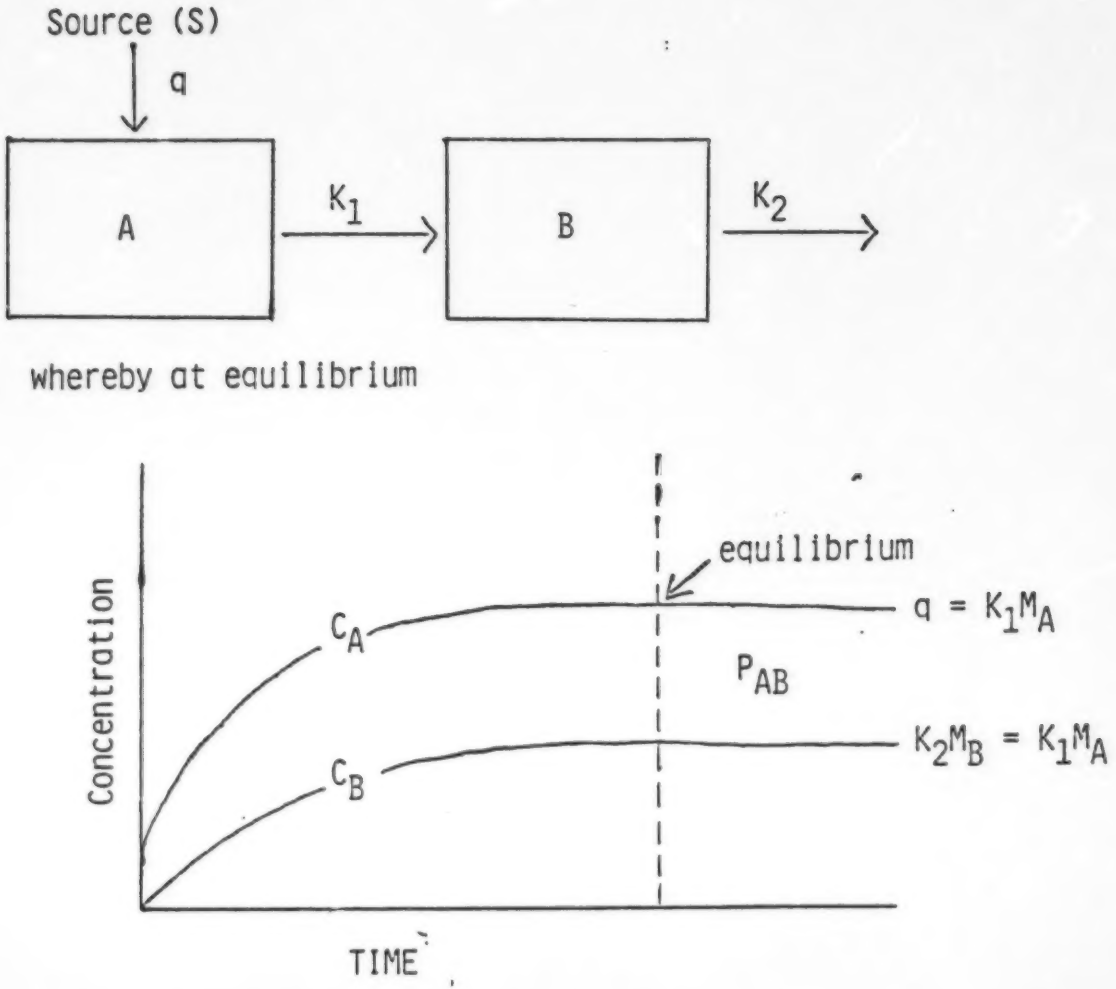


Figure 2. A simplified food chain model at equilibrium where C_A and C_B are pollutant concentrations in food chain compartments A and B, P_{AB} is an equilibrium concentration factor, K_1 and K_2 are transfer rate constants, M_A and M_B are compartment masses, and the system is exposed to a pollutant from a given source at a continuous flux q.

are corrected for the differences between the value obtained at equilibrium and the value attained at the time of harvest or at the end of a specified release period. Thus:

$$C_{B(t)} = C_A^* \times P_{AB} (1 - e^{-K_2 t}), \quad (2)$$

where

$C_{B(t)}$ = the concentration of a pollutant in food chain compartment B at time t subsequent to a continuous exposure to a pollutant,

K_2 = the effective first order rate constant for the loss of pollutants from food chain compartment B ($time^{-1}$), and

t = the time between initial exposure to the pollutant and harvest or the end of the release period.

When the reciprocal of the effective rate constant K_2^{-1} is very large with respect to t, equation (2) is simplified to

$$C_{B(t)} = C_A^* \times P_{AB} \times K_2 \times t. \quad (3)$$

When K_2^{-1} is very small with respect to t, equation (2) equals equation (1) and $CB(t)$ equals C_B^*. Quasi-equilibrium models can be reduced to a series of multiplicative chains, and calculations with these models can be readily performed by hand, or at least with the use of a hand calculator.

2.2.1.1 Derivation of Concentration Factors

Various methods are employed to derive equilibrium concentration factors (Barry 1979). The simplest method is to ratio pollutant concentrations in food chain compartments when these compartment concentrations are in equilibrium with each other. For example,

$$P_{AB} = C_B^*/C_A^*. \quad (4)$$

Other methods for determining concentration factors include the use of ratios of pollutant concentrations in food chain compartments averaged over sufficiently long time periods,

$$P_{AB} = \bar{C}_B/\bar{C}_A, \tag{5}$$

and ratios of infinite time-integrated concentrations obtained from acute exposure situations,

$$P_{AB} = IC_B/IC_A. \tag{6}$$

The infinite time-integrated concentrations IC_A and IC_B can be estimated using kinetic information on the rate constants for the transfer of a pollutant from a donor to a receptor food chain compartment and on the masses of these compartments (Barry 1979). Thus,

$$IC_A = \int_{-\infty}^{\infty} C_A(t)dt = \frac{Q_o}{(K_1 + K_3)M_A}, \tag{7}$$

$$IC_B = \int_{-\infty}^{\infty} C_B(t)dt = \frac{K_1 Q_o}{K_2 M_B (K_1 + K_3)}, \text{ and} \tag{8}$$

$$P_{AB} = \frac{K_1 M_A}{K_2 M_B}, \tag{9}$$

where the structure of the food chain is,

$$S \xrightarrow{Q_o} A \xrightarrow{K_1} B \xrightarrow{K_2} C,$$
$$\qquad\quad A \xrightarrow{K_3} D$$

K_1, K_2, and K_3 are first order rate constants, M_A and M_B are compartment masses, and the system is exposed to a single pulsed emission from source S of mass Q_o.

Experimentally, concentration factors are obtained most practically through either time averaging or time integrating measured concentrations in food chain compartments. Equilibrium or steady-state relationships are observed mainly in controlled laboratory studies, but are difficult to confirm under field conditions. Kinetic data, such as rate constants, are typically derived only from situations following either an acute release of a pollutant or an abrupt termination of a prolonged release. Examples of the derivation of concentration factors using various methods can be found in reports by Ng et al. (1977) and Saha (1969).

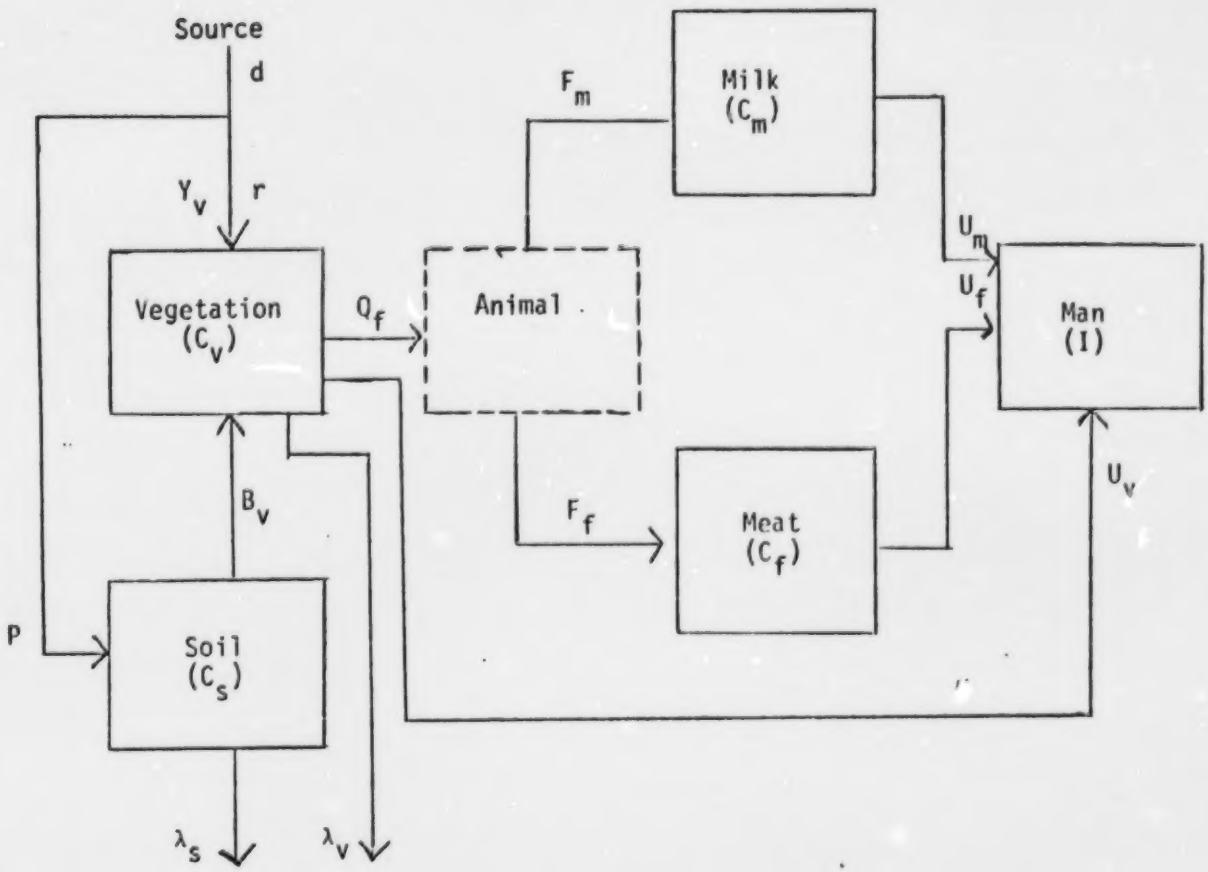
Uncertainties associated with the derivation of concentration factors from system kinetics are related to the potentially large temporal and spatial variability of rate constants obtained from acute contamination experiments. In situations where input and loss rate constants and compartment masses have been determined independently, there is also uncertainty introduced because of possible correlations among these parameters. In addition, equilibrium concentration factors are often derived from ratios of average concentrations of pollutants in soil, vegetation, and animal food products reported in the literature from unrelated references. For example, reported concentrations for a donor compartment of the food chain may be specific for completely different locations and time periods than are relevant for concentrations reported for receptor compartments of the food chain. Many food chain transfer factors listed for stable elements in quasi-equilibrium models developed for radiological assessments have been derived in this manner (USNRC 1977, Ng et al. 1978). The uncertainty associated with this method of parameter value derivation is also large because of the absence of correlations between donor and receptor compartments. When parameter values are so derived, it is easy to overlook situations that may result in significant bias in model predictions.

In the absence of data on the transfer of specific pollutants, concentration factors are often derived from collateral information on related substances having similar physical and chemical properties. The estimated behavior of long-lived radionuclides, for example, is often based on the known behavior of stable isotopes of the element. In addition to stable isotope analysis, chemical analogs are sometimes used to obtain an approximate estimate of transfer factors for radioactive and non-radioactive pollutants (Ng et al. 1977, Ng 1981, Baes et al. in preparation).

Most of the quasi-equilibrium models currently in use are similar in structure to those developed for the HERMES computer code for the assessment of radionuclide releases (Fletcher and Dotson 1971). At Oak Ridge National Laboratory, a similar model has been incorporated into the AIRDOS-EPA computer code (Moore et al. 1979) which combines food chain transport with atmospheric dispersion and dosimetry. Versions of this model have also been developed for the assessment of non-radioactive pollutants (Moghissi et al. 1980, Brenchley et al. 1977). In these models, equilibrium concentration factors have been generalized somewhat by implicitly expressing them as a function of numerous subparameters (Fig. 3).

2.1.1.2 Limitations of Quasi-equilibrium Models

Quasi-equilibrium food chain models have a limited scope of application. Predicted food chain concentrations are maximum or steady-state values resulting from a chronic, uniform release of pollutants into the environment. For non-uniform releases, quasi-equilibrium methods can be used if sufficiently long averaging times are selected (Barry 1979). For acute releases, the use of quasi-equilibrium models will result in estimates of time-integrated food concentrations, provided that average environmental conditions are prevailing at the time of discharge. Modifications of concentration factors will



Equations:

$$C_v = \frac{dr[1 - \exp(-\lambda_v t_v)]}{Y_v \lambda_v} + B_v C_s$$

$$C_s = \frac{d[1 - \exp(-\lambda_s t_s)]}{P\lambda_s}$$

$$C_m = C_v Q_f F_m$$

$$C_f = C_v Q_f F_f$$

$$I = (U_m C_m) + (U_c C_f) + (U_v C_v)$$

where:

C_v = the pollutant concentration in vegetation (µg/kg)

C_s = the pollutant concentration in soil (µg/kg)

C_m = the pollutant concentration in milk (µg/l)

C_f = the pollutant concentration in meat (µg/kg)

I = the intake rate of the pollutant by man via ingestion of food (kg or l/time)

d = the flux of the pollutant from a source (µg/m^2 ·time)

References proposing this model for pollutant assessment:

Fletcher and Dotson (1971)

Baker et al. (1976)

USNRC (1977)

Brenchley et al. (1977)

Moore et al. (1979)

McDowell-Boyer and Baes (1980)

Moghissi et al. (1980)

IAEA (1981)

r = the fraction of the flux intercepted by vegetation

Y_v = the vegetation biomass (kg/m^2)

λ_v = the removal rate of the pollutant from vegetation ($time^{-1}$)

t_v = the exposure period for vegetation (time)

B_v = the soil to plant concentration factor

p = the effective soil surface density ($\mu g/m^2$)

λ_s = the removal rate of the pollutant from the root zone of soil

t_s = the exposure period for soil

Q_f = the rate of vegetation ingestion by an animal (kg/time)

F_m = the milk to vegetation intake rate transfer coefficient (time/l)

F_f = the meat to vegetation intake rate transfer coefficient (time/kg)

U_m = the rate of milk consumption (l/time)

U_f = the rate of meat consumption (kg/time)

U_v = the rate of vegetation consumption (kg/time)

Figure 3. A commonly used version of a quasi-equilibrium model developed for the assessment of radionuclides and other pollutants.

be necessary, however, to account for conditions that may actually exist at the time of an acute release which may deviate considerably from the expected average (ICRP 1979). Nevertheless, in those circumstances where changes in the rate of exposure to pollutants is important for decisions, models must be developed to account for the dynamic behavior of pollutants. This requires data on the time-dependent rates of biophysical uptake, transfer, elimination, and chemical transformation of pollutants among the various interconnected compartments of terrestrial food chains. Quasi-equilibrium models are not suited for such calculations.

2.2.2 Dynamic Models

In order to improve the ability to predict the time dependency of pollutant transfer through terrestrial food chains, as well as to improve the state of knowledge about the flow of materials through ecological systems, recent emphasis has been on the development of dynamic models (Gillett et al. 1974, NRPB/CEA 1979, McDowell-Boyer et al. 1980). Dynamic models attempt to conceptually approximate the kinetics of a real system. The advantage of dynamic models is that they may be applied to any type of release (acute, intermittent, continuous or any combination of these).

In their simplest form, dynamic models are based on first-order kinetics with each input or loss term expressed as a rate constant multiplied by the concentration of a pollutant in a donor food-chain compartment. In this form, analytical solutions to the system of linear first-order differential equations are possible. However, as the expressions for individual transfers become more realistic, e.g., when the rate constants are replaced by time-dependent or concentration-dependent functions, analytical solutions are difficult to obtain. Numerical solution methods must then be called

upon to simulate the time behavior of pollutants in food chains. Since these methods require extensive calculations, the model in its most useful form becomes an algorithm coded for a computer.

Dynamic terrestrial food chain models have been developed as early as the late 1960's at Oak Ridge National Laboratory (Rohwer et al. 1969, Booth et al. 1971). These were intended for the assessment of radiological impact resulting from either an acute or a chronic release situation, and were basically donor controlled linear systems models (Fig. 4) composed of coupled first-order differential equations. Similar versions of this model have been developed for the terrestrial food chain assessment of DDT and DDE (O'Neill et al. 1971) and of cadmium (Walsh et al. 1977). The rate constants developed for the input and elimination of radionuclides from each food chain compartment of these models are time invariate.

The present state of development of dynamic food chain models for nuclear assessments emphasize the time dependence of the entire system to account for radionuclide daughter buildup with time, seasonal effects, and time-dependent rate constants requiring numerical solutions (Pleasant et al. 1980). Terrestrial food chain models having similar objectives are being developed in the United Kingdom (Simmonds et al. 1979). Dynamic food chain models for non-nuclear assessments are also being developed to account for pollutant dynamics during food chain transport and to simulate their behavior in the absence of empirical data derived rom field observations (BDM 1980).

Practically all food chain models developed at the process level for research purposes are dynamic as their objective is to mathematically simulate the behavior of pollutants in terrestrial systems as realistically as possible (Gillett et al. 1974). A linear model can be only an abstract conceptual approximation to the kinetics of a real system. In reality, terrestrial system kinetics will vary continuously over time as a function of

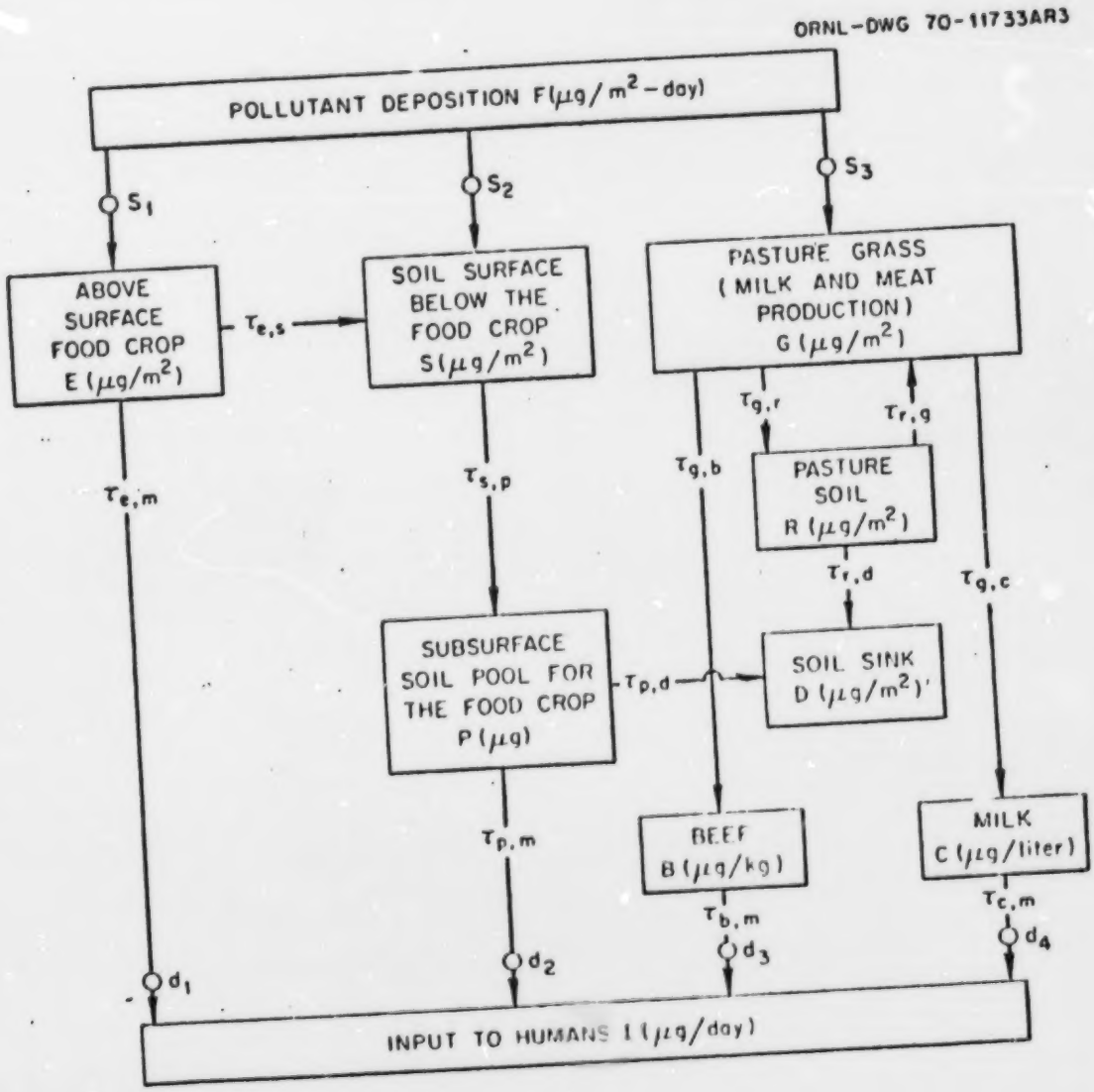


Figure 4. Block diagram of a donor controlled linear systems model for terrestrial food chains (Booth et al. 1971).

diurnal and seasonal changes in the physical environment as well as changes in the abundance, distribution, and composition of biota. In addition, terrestrial system kinetics may vary as a function of distance and direction from the point of release. The most realistic mathematical model, therefore, is a system of partial differential equations with coefficients varying as joint functions of time and location.

2.2.2.1 Limitations of Dynamic Models

The primary limitation associated with the development of dynamic models is the difficulty in obtaining appropriate data for quantifying model parameters. In most cases measurements have not been performed to determine the rate constants and transfer functions needed to simulate the dynamic behavior of pollutants in terrestrial food chains under temporally and spatially varying conditions. Even for simple linear systems models, values for rate constants must be ultimately determined from a time series of measurements. To obtain parameter values relevant to the assessment of acute releases, this time series of measurements must be repeated temporally and spatially. The experimental requirements to produce relevant dynamic transfer coefficients for even moderate-sized terrestrial food chain systems may be prohibitively large depending on how the model is to be applied (Miller 1978).

In the absence of kinetic data, input rates to compartments are sometimes derived from concentration factors and estimated elimination rates (Booth et al., 1971). For example, the uptake rate coefficient for milk of a cow, $r_{g,c}$ (m^2/l-day), was defined for the terrestrial food chain model depicted in Fig. 4 as

$$r_{g,c} = \frac{C_{eq}}{G_{eq}} \, r_{milk}, \qquad (10)$$

where

C_{eq}/G_{eq} = equilibrium nuclide concentration ratio of milk to grass, and

τ_{milk} = excretion rate from the udder of a milk-producing animal.

Thus, $\tau_{g,c}$ is constant for a constant elimination rate. This method of parameter derivation is subject to the same sources of uncertainties affecting derivation of the equilibrium concentration factor. In addition, values obtained for the concentration factor and excretion rate may not be representative of comparable conditions.

For dynamic models initially developed as research tools, the experimental determination of individual transfer coefficients for the assessment of a given pollutant may be impractical. This is because dynamic process-level models often are dependent on parameters that cannot be readily measured in the field without undue manipulation of the system. Also, with an increasing number of compartments or state variables within the model, there is an increasing need to measure all parameters simultaneously, otherwise important information on correlations between transfer coefficients will not be included in the model. Without specification of parameter covariance, increasing model complexity will increase the sensitivity of model calculations to error propagation (O'Neill et al. 1980, Gardner et al. 1980). The difficulties encountered in obtaining pollutant-specific data through simultaneous measurements for parameters used in a process-level model of the type illustrated in Figure 5 can be readily appreciated.

Because of the limitations associated with data acquisition for detailed dynamic process-level models, some have proposed restricting model development to readily measurable variables even though such variables may include an aggregation of several apparently distinct mechanisms. This proposal is

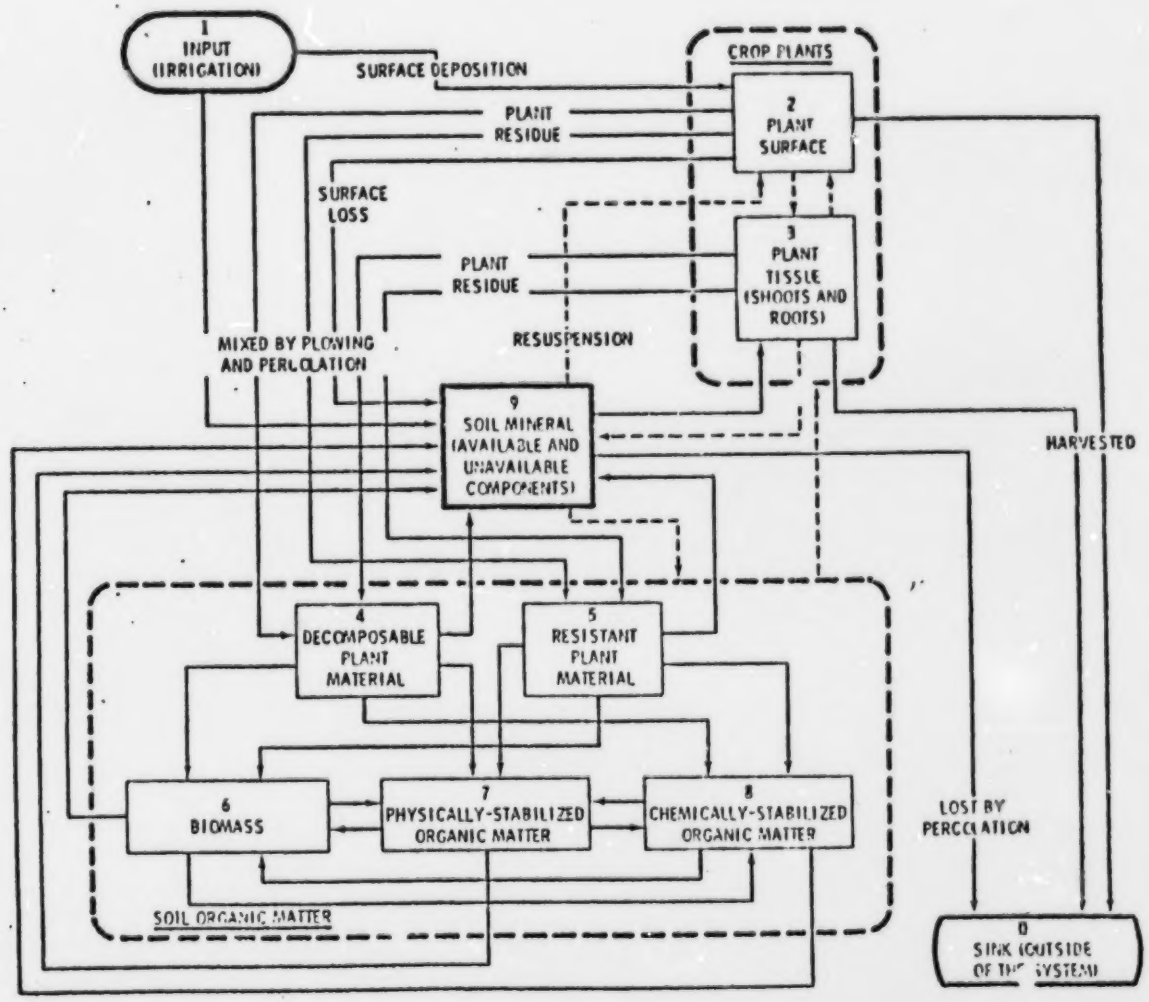


Figure 5. Compartmental model describing the long-term accumulation of radionuclides in crop plants (Schreckheise 1980).

based on the argument that *at best, any model can only hope to be an abstraction of reality.* Therefore, when the formal structure of the model is reduced to include only the flow of pollutants between readily measurable major compartments of the terrestrial system, correlations among the various processes will be included implicitly in parameter measurements. This should improve the predictive capability of the model for situations similar to those in which parameters have been measured. Enhancing the ability to measure parameters under field conditions should also improve the ability to field test or validate model predictions which is a most desirable step in determining the true utility of assessment models (Miller et al. 1980, Hoffman 1980).

3. APPLICATION OF TERRESTRIAL FOOD CHAIN MODELS IN ASSESSMENTS

3.1 Screening for Important Pollutants and Pathways

Realizing that quantified predictions are based on simplifying assumptions and generalizations of more complex real-world systems, a useful application of terrestrial food chain assessment models is to screen for potentially important pollutants and pathways. Screening is usually performed as a part of a preliminary assessment. Typically it involves comparison of conservatively biased model predictions with established acceptable limits of pollutant concentrations in foods or exposures or doses to man. Without the establishment of limiting values for acceptable pollutant concentrations in terrestrial food chains, the process of screening becomes meaningless.

Conservatism is employed to decrease the probability that model predictions will result in an underestimation of actual food chain concentrations.

Thus, when a predicted concentration is below a certain limit, the pollutant and pathway usually can be considered of negligible importance (Dacre et al. 1980, Hoffman and Kaye 1976). However, pollutants and pathways predicted to result in food chain concentrations that encroach upon or exceed limiting values can only be designated as *potentially* important until further analysis of model assumptions and existing data or additional research can be conducted to improve the accuracy of model predictions.

Conservatism for screening purposes may be assigned to the model structure, the selection of parameter values, and/or the application of a safety factor to either model predictions or established limits (Dacre et al. 1980). The amount of and type conservatism applied for screening will be dependent on the level of understanding of the pollutant in terrestrial food chains, the quality of available data, and the history of model validation. The degree of conservatism will also be related to the actual or perceived health risk associated with potentially exceeding an established limit. Thus, the greater the level of knowledge about the pollutant in a specific terrestrial system and the lower the health risk associated with exceeding a limit, the lesser is the amount of conservatism required for screening (Fig. 6).

Model validation studies will be useful to determine the extent of the conservative bias in model predictions and to identify possible situations where potential underestimation can be avoided. Once model validation has determined the degree of overestimation associated with conservative bias, efforts can be directed at increasing the accuracy of model predictions. Model validation of screening models will also serve to place an upper bound on predictive uncertainty. Thus, if validation indicates that a screening model does not overpredict by more than a certain quantity, predicted food chain concentrations exceeding limits by more than this amount would be

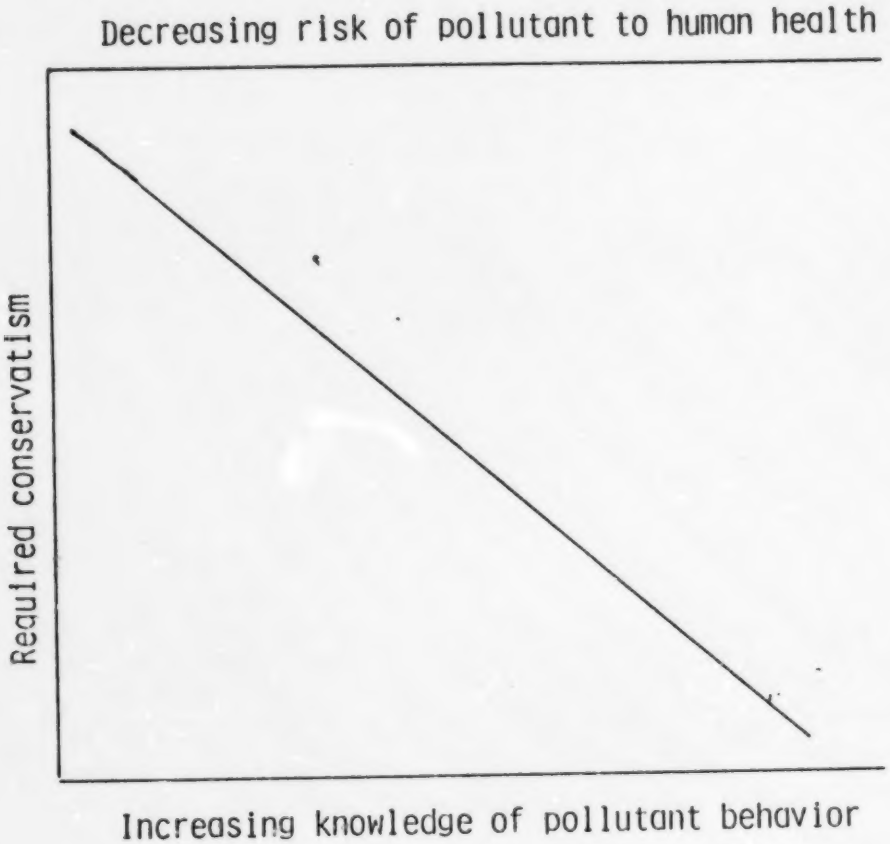


Figure 6. Relationship between the degree of conservatism required for screening calculations, the level of knowledge of the behavior of the pollutant in terrestrial food chains, and the risk of the pollutant to human health.

immediately implicated as unacceptable. Without validation, further research and evaluation would be required prior to the final designation of unacceptable food chain concentrations.

Sometimes process-level models are proposed for screening calculations. These situations most frequently occur when the actual behavior of the pollutant in terrestrial food chains is not well understood and when relevant parameter values and model validation results are lacking. The presence of a complex mathematical structure in such a situation presents an aura of model realism that may appear to produce more defensible results than models having a less complex structure (Hoffman et al. 1978a). However, unless available data and validation results can justify the need for increased complexity for screening purposes, the assumption of increased defensibility with increased structural complexity may be unwarranted.

Screening models associated with a sufficient degree of conservatism will be most useful for calculations performed for pollutants, pathways, and locations that do not have an extensively developed base of data. The use of such models to evaluate the significance of pollutants released from industrial and agricultural operations in developing countries will enable an efficient identification of situations requiring more detailed analyses and expenditures of resources to improve the accuracy of preliminary predictions. Even in the United States of America, screening models are essential for evaluating potentially hazardous chemicals under the Toxic Substances Control Act of 1976 (Muir 1980) because data on the toxicity and behavior of each of the thousands of specific chemical species that must be considered are simply not available.

For the assessment of radionuclides, a quasi-equilibrium model using generic default transfer factors is being published by the International Atomic Energy Agency for use as a tool in screening for radioisotopes and

pathways resulting in doses to critical groups of the population that approach pre-selected dose limits within one order of magnitude (IAEA 1981). The development of this model was specifically intended for use by developing countries that may not have access to relevant data for use in assessment calculations. For screening of other pollutants of potential concern to human health, Dacre et al. (1980) propose the application of models utilizing a series of equilibrium concentration factors.

Although all types of models used for assessment and research purposes can be adapted for screening calculations, it is our experience that models are most successful for screening when they are easily implemented by individuals not directly related to their development. Usually, calculational procedures are not extensive and the data base includes generic default parameter values for use in the absence of site-specific information. A crude calculation will be entirely acceptable for screening purposes, if resultant concentrations are well below established limits.

3.2 The Use of Models for Optimization of Releases

Another application of assessment models is to optimize pollutant release rates to levels considered as low as reasonably achievable. Optimization, therefore, involves a comparison between the benefit gained in terms of reduced health risk and the societal cost required to achieve this reduced risk (ICRP 1977). Terrestrial food chain models developed for optimization calculations would be used to justify the costs encountered to reduce pollutant releases and to compare health risks related to pollutants discharged from alternative types of industrial or agricultural practices.

Screening models have limited applicability to optimization calculations because conservative overestimates of pollutant concentrations can

grossly mislead decisions resulting in potentially unwarranted financial costs or unnecessary dismissal of potentially viable technologies. For optimization, unbiased predictions are preferable to conservative estimates. The achievement of unbiased model predictions, however, requires knowledge of predictive uncertainties. In the absence of such knowledge, models developed for optimization often employ "best estimate" or average values for parameters derived using the judgment of the model developer or the model user. These judgment estimates also may result in misleading conclusions without some indication about the possible uncertainties, because best estimate values may incorrectly predict actual conditions prevailing in a given assessment situation.

Uncertainties associated with model predictions are best determined through model validation studies (Holling 1978, Schwarz 1980, Shaeffer 1980). Unfortunately, only limited validation for a few pathways and radioisotopes have been performed for radiological assessment models (Hoffman 1980, Erb 1979, Haywood et al. 1980), with no known validation having been performed for the assessment of non-nuclear pollutants. Nevertheless, the extent of model validation required before models can be reliably used will be directly related to the degree of accuracy expected of model predictions (Fig. 7). Therefore, present optimization evaluations are limited by uncertainties associated with the judgment used to derive "best estimate" or "expected value" predictions for specific pollutants and food chain pathways.

Sometimes, in the absence of model validation, process-level models are proposed for optimization calculations because of the apparent realism in model structure. Again, as with the application of such models for screening, the usefulness of process-level models is limited because of their complexity, the magnitude of data requirements, and sensitivity to error propagation.

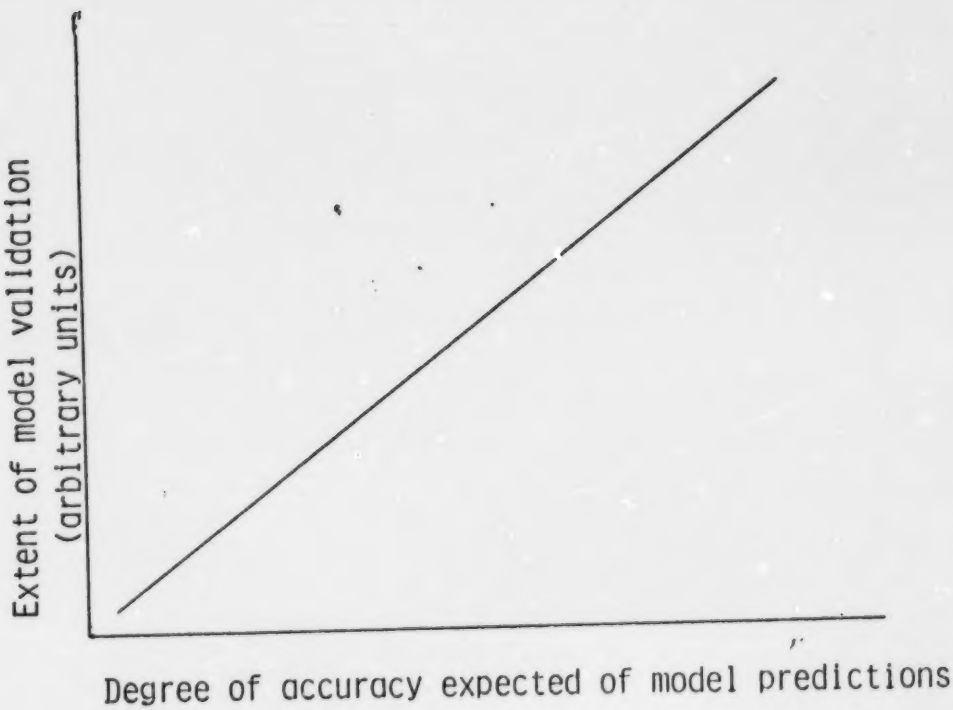


Figure 7. Relationship between model validation and the degree of accuracy expected of models used for optimization calculations.

Furthermore, as discussed previously, validation of these models is difficult or impractical. Given the current state of model validation, predictions made with process-level models serve to provide a formalized viewpoint about the possible behavior of pollutants in terrestrial systems that at least can be compared with the results of other types of model formulations.

4. DIRECTIONS FOR IMPROVEMENT

4.1 Quantification and Reduction of Predictive Uncertainties

Emphasis on future model improvements for the assessment of pollutant transport in food chains is being placed on the quantification and reduction of predictive uncertainty (Hoffman et al. 1978b, Little and Miller 1979, Miller et al. 1980). The use of screening models will facilitate identification of pollutants and pathways requiring models with improved predictive accuracy. It is our contention that only through more agressive efforts to validate terrestrial food chain models will desired improvements in predictive accuracy be realized.

Model validation represents actual field testing under the range of conditions that a model may be intended for use (Holling 1978, Schwarz 1980). Model validation will therefore be expensive and time consuming. The trade-off between an agressive validation program and no validation is the difference between the cost required to identify model uncertainties and the risks involved with predictions whose uncertainties are either unknown or grossly over-conservative.

In the absence of results from model validation, some investigators have attempted to analyze predictive uncertainties using analytical and numerical techniques for simulating the propagation of error in terrestrial

food chain model predictions due to the variable nature of model parameters (Hoffman and Baes 1979, McKay et al. 1979, Schwarz and Hoffman 1981, O'Neill et al. 1981, Matthies et al. 1981). This approach enables a model to produce a range of predictions with a distinct frequency distribution rather than just a single value. Figure 8 illustrates an example of such an error propagation analysis for the transport of ^{131}I over the pasture-cow-milk pathway. Error propagation techniques by themselves, however, can only provide an estimate of predictive imprecision (Schwarz 1980). The departure of predicted values from real-world concentrations of pollutants in terrestrial foods remains unknown without field testing, although it is conjectured that real-world values should be encompassed by the range of predictions produced through error propagation (ICRP 1979).

For quantifying predictive uncertainty, model validation offers the possibility to identify both bias and imprecision when combined with error propagation techniques. Reduction in predictive uncertainty can be accomplished through model validation by calibrating model predictions with observations. Calibration of predictions with observations is most justified when there is a high correlation between predictions and observations. A poor correlation between predictions and observations indicates the need for improvement of model structure or in the determination of key parameters. However, even when poor correlations predominate, model predictions may still undergo calibration to assure a sufficient margin of conservatism for screening calculations. Nevertheless, such models should only be applied within the realm of environmental and ecological conditions tested through validation studies. Extrapolation of model predictions beyond this realm is extremely dangerous when predictions exhibit poor correlations with observations, especially when the bottom line is a certain degree of health risk.

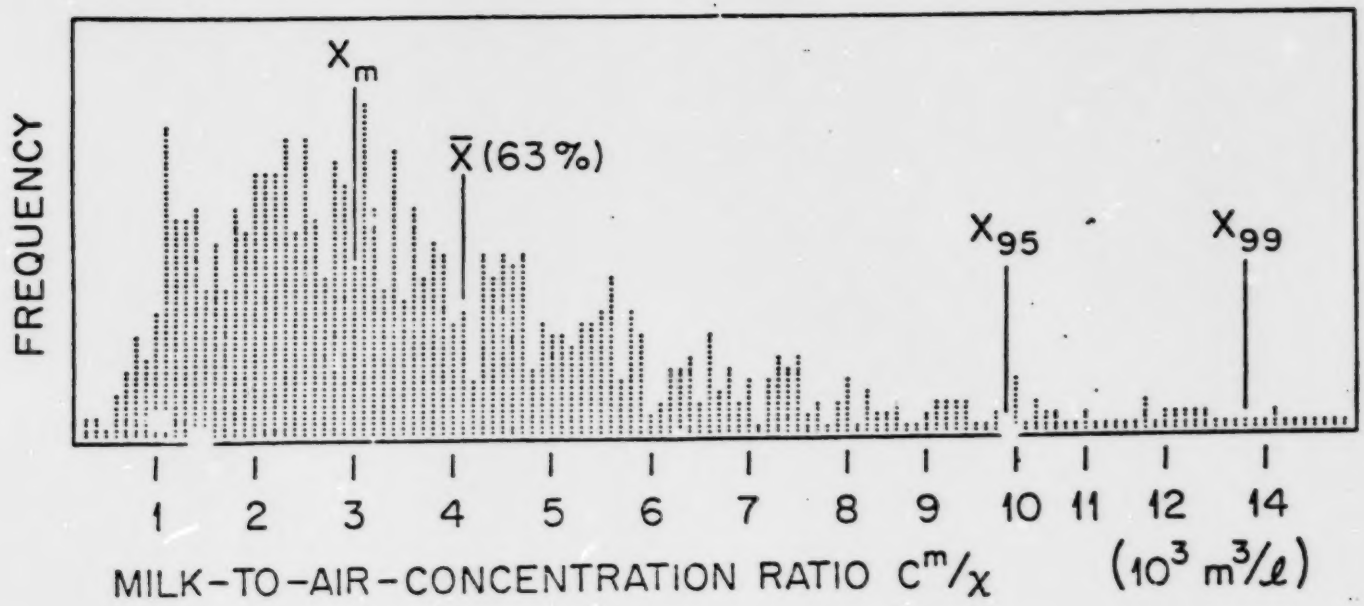


Figure 8. The milk-to-air concentration ratio C^m/χ for $^{131}I_2$ simulated using Monte Carlo computations with estimates of the median (X_m), mean ($\bar{X}$), 95th (X_{95}), and 99th (X_{99}) percentiles included within the distribution of predicted values (Schwarz and Hoffman 1981).

4.2 Improve Site-Specific Assessment Capability

Further reduction in predictive uncertainty can be expected with improved capability for site-specific assessments. Current terrestrial food chain models contain parameters whose values may be readily determined for a specific site, such as standing vegetation biomass, the bulk density of surface soil, and some aspects of cattle management practices (USNRC 1977, Moghissi 1980, IAEA 1981). But in general, pollutant-specific transfer factors or rate constants cannot be determined for a specific site without extensive monitoring or experimental investigation. At present these transfer factors or rate constants have been derived from the global literature and aggregated for major food categories such as vegetables, milk, and meat. The variation of these parameters with plant and animal species or breeds, agricultural practices, soil characteristics, and climatic regimes has not been determined for most types of pollutants.

A considerable amount of site-specific experimentation can be avoided if it is possible to correlate pollutant-specific parameters with easily measurable environmental variables such as soil pH, temperature, biomass, etc. For radionuclide assessments, correlations with a few environmental variables have been developed for isotopes of strontium and cesium (Bieshold et al. 1979, Hoffman 1980). However, development of data to permit derivation of such relationships requires a large investment in specifically designed laboratory and field research.

Improving the site-specific capability of assessment models will also require improved knowledge of human dietary habits for specific locations. Currently, health risks are typically assessed by combining predicted food concentrations with rates of food consumption by humans corresponding to reference diets assumed to represent average or maximally exposed individuals

(ICRP 1975, USNRC 1977). The relevancy of these assumed diets to specific situations may contribute to assessment uncertainty; however, determination of regional-specific or locational-specific dietary habits may be difficult because of rapidly changing trends in future market practices (Rupp 1980).

4.3 Consideration of Model Simplification

An additional consideration for future improvement of terrestrial food chain assessment models for assessment purposes is the achievement of an optimum level of model simplification. Usually, the "best" model for a given assessment will be the model that is easiest to use and which produces results within an acceptable degree of accuracy which address the questions asked of the model by the model user (Shaeffer 1980). The degree of "acceptable accuracy" will be determined by the results of validation studies and the risks associated with over or underestimating the impact to human health.

Simplification should be most achievable for models used for screening calculations because of inherent conservatism. However, if an extensive series of validation tests have been performed, it may also be possible to simplify models developed for optimization calculations through calibration of model predictions to observations. For most situations within the realm of our experience, many complex model formulations appear to be more suited as research rather than assessment tools because of deficiencies in available data and difficulties involved with achieving further improvements in the data base. Nevertheless, these models may still have utility for guiding experimental studies and obtaining valuable insights on pollutant behavior and potential pollutant bioaccumulation.

Additional structural complexity inevitably will be required to increase the flexibility of terrestrial food chain models for site-specific assessments.

For the sake of practicality, however, it may be advisable to maintain structural simplicity with the dependencies of parameters on site-specific and pollutant-specific characteristics being considered external to the model that is used for assessments. Thus, simple transfer factors or rate constants may actually be derived from tables of data and more complex model formulations that are kept separate from the basic assessment model. Site-specific models consisting of few parameters for important pollutants and pathways should be more amenable than more complex formulations to validation studies conducted under conditions of high temporal and spatial variability because of reduced demands on simulataneous parameter measurement (Hoffman 1980). For assessment purposes, an optimum level of structural complexity can be envisioned when all parameters are readily measurable and the possibility of predictive error due to unforeseen correlations among the parameters is small. The optimum level of simplification, however, can only be determined on the basis of results gained through model validation and the ease with which the model can be implemented.

5. CONCLUSIONS

The primary purpose for the development and application of terrestrial food chain models in the assessment of pollutant releases to the environment is the prediction of human health impact. Most of the examples we have used in this paper have been taken from models applied for the assessment of radionuclides because formal assessment methodologies for other types of pollutants are still at an early stage of development. Models used in assessments have been designed to produce quasi-equilibrium or dynamic predictions; however, both model types suffer from limitations in parameter value determination and validation. Because the determination of parameter

values is highly dependent on empirical derivation, extrapolation of model predictions to locations, time periods, and chemical species of pollutants for which data are not available must be exercised with due caution. Furthermore, model predictions are inherently limited by the information available for determination of model structure and parameter values. A model cannot be expected to provide more information than was put into it. The limitations in parameter quantification and model validation will therefore probably restrict most uses of terrestrial food chain models in assessments to a type of screening calculation. The potential for unexpected misprediction exists each time a model is applied to a new situation, regardless of the results of past validation experiments. Therefore, there will always be a need to implement environmental monitoring programs to ensure that potential model misprediction does not result in unacceptable consequences once releases of pollutants to terrestrial food chains actually occur.

6. ACKNOWLEDGMENTS

The authors are greatly indebted to Keith F. Eckerman, Charles T. Garten, Jr., Joseph K. Soldat, John R. Trabalka, and E. Ivan White for the provision of key references related to the development and application of models for the prediction of pollutant transport in terrestrial food chains. We are also greatly indebted to Robin L. Smith for her assistance in preparation of this manuscript.

7. REFERENCES

Baes, C. F., III, R. D. Sharp, A. L. Sjoreen, and R. W. Shor (In preparation). *A Review and Analysis of Parameters for Assessing Transport of Environmentally Released Radionuclides through Agriculture.* ORNL-5786.

Baker, D. A., G. R. Hoenes, and J. K. Soldat, 1976. *FOOD — An Interactive Code to Calculate Internal Radiation Doses from Contaminated Food Products.* BNWL-SA-5523.

Barry, P. J., 1979. *An Introduction to the Exposure Commitment Concept with Reference to Environmental Mercury.* MARC report No. 12. Chelsea College, University of London.

BDM Corporation, 1980 (draft report). *Sewage Sludge Pathogen Transport Model Project — Final Report. Vol. I.* Health Effects Research Laboratory, U. S. Environmental Protection Agency, Cincinnati, Ohio. BDM/TAC-79-528-TR.

Biesold, H., P. Gruner, and K. Heim, 1978. Ermittlung von Transferfaktoren Boden/Vegetation anhand standortspezifischer Bodenparameter, in: *Radioaktivität und Umwelt,* I:482-509, Fachverband für Strahlenschutz e.V.

Booth, R. S., S. V. Kaye, and P. S. Rohwer, 1971. Systems analysis methodology for predicting dose to man from a radioactively contaminated terrestrial environment, in: *Radionuclides in Ecosystems,* 3rd National Symposium on Radioecology, Oak Ridge 1971, USAEC CONF 710501-P2.

Brenchley, D. L., J. K. Soldat, J. A. McNeese, and E. C. Watson, 1977. *Environmental Assessment Methodology for the Nuclear Fuel Cycle.* BNWL-2219.

Dacre, J. C., D. H. Rosenblatt, and D. R. Cogley, 1980. Preliminary pollutant limit values for human health effects. *Environ. Sci. and Tech.* 14:778-784.

Edwards, C. A. (ed.) 1973. *Environmental Pollution by Pesticides.* Plenum Press, London.

Erb, J. C., 1979. An assessment of the environmental transport of radioiodine in the air-grass-cow-milk pathway using reported environmental monitoring data. *Proceedings of Health Physics Society Twelfth Midyear Topical Symposium,* February 11-15, 1979, Williamsburg, Virginia, pp. 504-514.

Fletcher, J. F. and W. L. Dotson, 1971. *HERMES — A Digital Computer Code for Estimating Regional Radiological Effects from the Nuclear Power Industry.* HEDL-TM-71-168.

Gardner, R. H., R. V. O'Neill, J. B. Mankin, and D. Kumar, 1980. Comparative error analysis of six predator-prey models. *Ecology* 61:323-332.

Garner, R. J., 1971. Transfer of radioactive materials from the terrestrial environment to animals and man. *CRC Crit. Rev. Environ. Control* 2:337-385.

Garten, C. T., Jr., 1978. A review of parameter values used to assess the transport of plutonium, uranium and thorium in terrestrial food chains. *Environ. Res.* 17:437-452.

Gillett, J. W., J. Hill IV, A. W. Jarvinen, and W. P. Schoor, 1974. *A Conceptual Model for the Movement of Pesticides Through the Environment.* EPA-660/3-74-024.

Haywood, S. M., J. R. Simmonds, and G. S. Linsley, 1980. *The Development of Models for the Transfer of ^{137}Cs and ^{90}Sr in the Pasture-Cow-Milk Pathway using Fallout Data.* NRPB-R110.

Hoffman, F. O., 1980. Terrestrial food chains, in: *Accuracy in Dose Calculations for Radionuclides Released to the Environment* (K. H. Lindakers and H. Bonnenberg eds.), GUW, Aldenhoven.

Hoffman, F. O. and C. F. Baes III (eds.), 1979. *A Statistical Analysis of Selected Parameters for Predicting Food Chain Transport and Internal Dose of Radionuclides.* ORNL/NUREG/TM-282.

Hoffman, F. O. and S. V. Kaye, 1976. Terrestrial exposure pathways: potential exposures of man from the environmental transport of waste nuclides, in: *Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle.* Denver, Colorado, July 11-16, 1976, U. S. Energy Research and Development Administration Report CONF-76-0701, pp. 524-538.

Hoffman, F. O., D. L. Shaeffer, C. W. Miller, and C. T. Garten, Jr., 1978a. *Proceedings of a Workshop on the Evaluation of Models used for the Environmental Assessment of Radionuclide Releases.* U. S. Department of Energy Report CONF-770901.

Hoffman, F. O., D. L. Shaeffer, C. F. Baes III, C. A. Little, C. W. Miller, D. E. Dunning, Jr., E. M. Rupp, and R. W. Shor, 1978b. An evaluation of uncertainties in radioecological models, in: *Radioaktivität und Umwelt,* II:610-648, Fachverband für Strahlenschutz e.V.

Holling, C. S. (ed.), 1978. *Adaptive Environmental Assessment and Management.* John Wiley and Sons, Chichester.

International Atomic Energy Agency, 1981. *Generic Models and Parameters for Assessing the Environmental Transfer of Radionuclides in Predicting Exposures to Critical Groups from Routine Releases.* Vienna (publication in press).

International Commission on Radiological Protection, 1975. *Report of the Task Group on Reference Man.* ICRP Publication 23, New York, Pergamon Press.

International Commission on Radiological Protection, 1977. *Recommendations of the International Commission on Radiological Protection.* ICRP Publication 26, New York, Pergamon Press.

International Commission on Radiological Protection, 1979. *Radionuclide Release into the Environment: Assessment of Doses to Man.* ICRP Publication 29, New York, Pergamon Press.

Lichtenstein, E. P., 1972. Environmental factors affecting fate of pesticides, in: *Degradation of Synthetic Organic Molecules in the Biosphere,* National Academy of Sciences, Washington, D.C., pp. 190-205.

Little, C. A. and C. W. Miller, 1979. *The Uncertainty Associated with Selected Environmental Transport Models.* ORNL-5528.

Matthies, M., K. Eisfeld, H. Paretzke, and E. Wirth, 1981. Stochastic calculations for radiation risk assessment: a Monte-Carlo approach to the simulation of radiocesium transport in the pasture-cow-milk food chain. *Health Physics* 40:764-770.

McDowell-Boyer, L. M. and C. F. Baes, III, 1980. Terrestrial food chain transport, in: *Recommendations Concerning Models and Parameters Best Suited to Breeder Reactor Environmental Radiological Assessments,* pp. 23-54, ORNL-5529.

McDowell-Boyer, L. M., G. G. Killough, and J. C. Pleasant, 1980. Dynamic modeling of radionuclides in terrestrial food chains. *Trans. Am. Nucl. Soc.* 34:85-86.

McKay, M. K., W. J. Conover, and R. J. Beckman, 1979. A comparison of three methods for selecting values of input variables in the analysis of output from a computer code, *Technometrics* 21(2):239-245.

Miller, C. W., D. E. Dunning, Jr., E. L. Etnier, D. C. Kocher, L. M. McDowell-Boyer, H. R. Meyer, and P. S. Rohwer, 1980. *Recommendations Concerning Research and Model Evaluation Needs to Support Breeder Reactor Environmental Radiological Assessments.* ORNL/TM-7491.

Miller, D. R., 1978. Models for total transport, in: *Principles of Ecotoxicology* (G. C. Butler ed.), pp. 71-90. John Wiley and Sons, Chichester.

Moghissi, A. A., R. E. Marland, F. J. Congel, and K. F. Eckerman, 1980. Methodology for environmental human exposure and health risk assessment, in: *Dynamics, Exposure and Hazard Assessment of Toxic Chemicals* (R. Haque ed.), Ann Arbor Science, Ann Arbor, Michigan, pp. 471-490.

Moore, R. E., C. F. Baes, III, L. M. McDowell-Boyer, A. P. Watson, F. O. Hoffman, J. C. Pleasant, and C. W. Miller, 1979. *AIRDOS-EPA: A Computerized Methodology for Estimating Environmental Concentrations and Dose to Man from Atmospheric Releases of Radionuclides.* EPA 520/1-79-009.

Muir, W. R., 1980. Chemical selection and evaluation: Implementing the toxic substances control act, in: *Dynamics, Exposure and Hazard Assessment of Toxic Chemicals in the Environment* (R. Haque ed.), Ann Arbor Science, Ann Arbor, Michigan, pp. 15-20.

National Radiological Protection Board/Commissariat a l'Energie Atomique, 1979. Methodology for evaluating the radiological consequences of radioactive effluents released in normal operations, CEC report V/3865/79e.

Ng, Y. C., C. S. Colsher, D. J. Quinn, and S. E. Thompson, 1977. *Transfer Coefficients for the Prediction of the Dose to Man via the Forage-Cow-Milk Pathway from Radionuclides Released to the Biosphere.* UCRL-51939.

Ng, Y. C., C. S. Colsher, and S. E. Thompson, 1978. Transfer coefficients for terrestrial food chains — their derivation and limitations, in: *Radioaktivität und Umwelt,* I:455-481, Fachverband für Strahlenschutz e.V.

Ng, Y. C., 1981. A review of transfer factors for assessing the dose from radionuclides in agricultural products. UCRL-85138, Revision 1, and *Nuclear Safety* (in press).

O'Neill, R. V., O. W. Burke, and R. S. Booth, 1971. *A Simple Systems Model for DDT and DDE Movement in Human Foodchains.* ORNL/IBP-71/9.

O'Neill, R. V., R. H. Gardner, F. O. Hoffman, and G. Schwarz, 1981. Parameter uncertainty and estimated radiological dose to man from atmospheric ^{131}I releases: A Monte Carlo approach. *Health Physics* 40:760-764.

O'Neill, R. V., R. H. Gardner, and J. B. Mankin, 1980. Analysis of parameter error in a nonlinear model. *Ecological Modeling* 8:297-311.

Pleasant, J. C., L. M. McDowell-Boyer, and G. G. Killough, 1980. *RAGTIME: A Fortran IV Implementation of a Time Dependent Model for Radionuclides in Agricultural Systems.* ORNL/NUREG/TM-371.

Rohwer, P. S., S. V. Kaye, K. E. Cowser, and W. S. Snyder, 1969. Predicting radiation dose equivalents for populations I. Dose models and methods of application. *Bio. Sci.* 19:238-241.

Rupp, E. M., 1980. Age dependent values of dietary intake for assessing human exposures to environmental pollutants. *Health Physics* 39:151-163.

Russell, R. S., 1966. Entry of radioactive materials into plants, in: *Radioactivity and Human Diet* (R. S. Russell ed.), Pergamon Press, Oxford.

Saha, J. G., 1969. Significance of organochlorine insecticide residues in fresh plants as possible contaminants of milk and beef products. *Residue Rev.* 26:89-127.

Schreckhise, R. G., 1980. *Simulation of the Long-Term Accumulation of Radio-contaminants in Crop Plants.* PNL-2636.

Schwarz, G., 1980. General aspects of accuracy in dose calculations, in: *Accuracy in Dose Calculations for Radionuclides Released to the Environment* (K. H. Lindakers and H. Bonnenberg eds.), GUW, Aldenhoven.

Schwarz, G. and F. O. Hoffman, 1981. Imprecision of dose predictions for radionuclides released to the environment: an application of a Monte-Carlo simulation technique. *Environ. International* (in press).

Shaeffer, D. L., 1980. A model evaluation methodology applicable to environmental assessment models. *Ecological Modeling* 8:275-295.

Simmonds, J. R., G. S. Linsley, and J. A. Jones, 1979. *A General Model for the Transfer of Radioactive Materials in Terrestrial Food Chains.* NRPB report R89.

U. S. Nuclear Regulatory Commission, 1977. Calculation of annual doses to man from routine releases of reactor effluents for the purpose of evaluating compliance with 10 CFR 50, Appendix I. *Regulatory Guide 1.109.* Revision 1, October (1977).

Walsh, P. J., G. G. Killough, D. C. Parzyck, P. S. Rohwer, E. M. Rupp, B. L. Whitfield, R. S. Booth, and R. J. Raridon, 1977. *A Cumulative Hazard Index for Assessing Limiting Exposures to Environmental Pollutants.* ORNL-5263.